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Technical Report

RESISTANCE HOT PRESSING

U. S. Office of Naval Research
Metallurgy Branch

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I. INTRODUCTION

Following World War II a research group at Alfred University, under contract with the Office of Naval Research, commenced a fundamental study of new materials for high stress, high temperature applications¹. In the course of this research, it became necessary to produce dense specimens of many varied compositions. Some specimens were formed by vacuum-sintering of cold-pressed compacts; some were hot-pressed using an induction hot-press furnace. The limitations of these methods, regarding this investigation, soon became apparent.

Sintering was suitable for many oxides, but was not applicable to carbides because of their tendency to decompose. The achievement of maximum density by this method was uncertain. Many compositions could not be sintered because of the broad range in melting points of the components; the low-melting materials would tend to melt out of the compact. The inherent slow heating and cooling rates encountered were a great disadvantage, in view of the large number of specimens to be prepared.

Induction hot pressing proved to be inordinately time consuming. Temperature measurement and control were subject to rather large error. Loading and unloading the furnace was objectionably messy because of the powdered carbon required to maintain the necessary close coupling and to provide a high degree of thermal insulation. Heating rates of certain components were greatly affected because of their differing responses to eddy current and hysteresis loss heating effects produced by the induction coil.

In view of the large number of compositions to be explored, it was imperative that a rapid fabrication method be devised. Such a technique should produce dense specimens of varied composition without the difficulties encountered with vacuum-sintering and induction hot pressing. A resistance hot-pressing method was developed to meet this need. Most of the test specimens were formed by this procedure.

The success of the original resistance hot press led to its ultimate "upscaleing" to produce larger specimens required for another research program. In time, a much larger resistance hot press was developed to permit fabrication of specimens of many different shapes.

II. DEVELOPMENT OF RESISTANCE HOT-PRESSING FURNACES

A. Jacketed Resistance Hot Press

Many of the materials to be studied under the O. N. R. program¹ were quite expensive to obtain. In order to prepare the full series of proposed compositions incorporating these expensive materials, small specimen size was indicated. The physical properties testing program was planned, therefore, to utilize small cylindrical specimens, 1 cm. in diameter by 1 cm. in length, for most of the determinations. On this basis, the resistance hot press² was designed to fabricate such specimens rapidly.

For simplicity of operation, the die assembly was used also as the resistance heating element. Graphite was selected as the construction material for these parts because of its easy machinability, its good strength at elevated temperatures, and its resistivity characteristics. Fig. 1 shows two views of

the graphite dies: the view at the left shows a portion of the central cross-section of a loaded die ready for hot pressing. The die at the right, in exploded view, illustrates all of the graphite parts in their proper relationship.

The die shown in Fig. 1 incorporates many improvements in design, as developed after much experimentation. The pushrod was made with an enlarged top section to insure good electrical contact with the furnace plunger, and to facilitate die alignment. The upper and lower shims were necessary to permit easy disassembly of the die in the event of extrusion of the sample and/or reaction with the graphite. The enlarged ends of the graphite sleeve provided the necessary mechanical strength, and the reduced cross-section of the central portion served to concentrate the dissipation of electrical energy as thermal energy in the region surrounding the specimen, thereby developing high temperatures at very rapid rates. It was essential that the cross-sectional area through the high-resistance portion of the sleeve be smaller than the cross-sectional area of the pushrod to insure concentration of heating at the specimen location. The pedestal was made as large as practicable to provide for die alignment and to effect good electrical contact with the furnace base. The "necked-down" portion of the pedestal was devised to function as a thermal barrier to limit heat loss from the specimen; in this regard, it appears feasible to utilize some carbon parts in the pedestal construction to effect a thermal barrier and thereby promote more uniform heating throughout the specimen.

In order to prevent catastrophic oxidation of the die parts during heating, the furnace was constructed with a water-cooled jacket having fittings by which to flush the chamber with an inert gas, such as Argon or Helium.

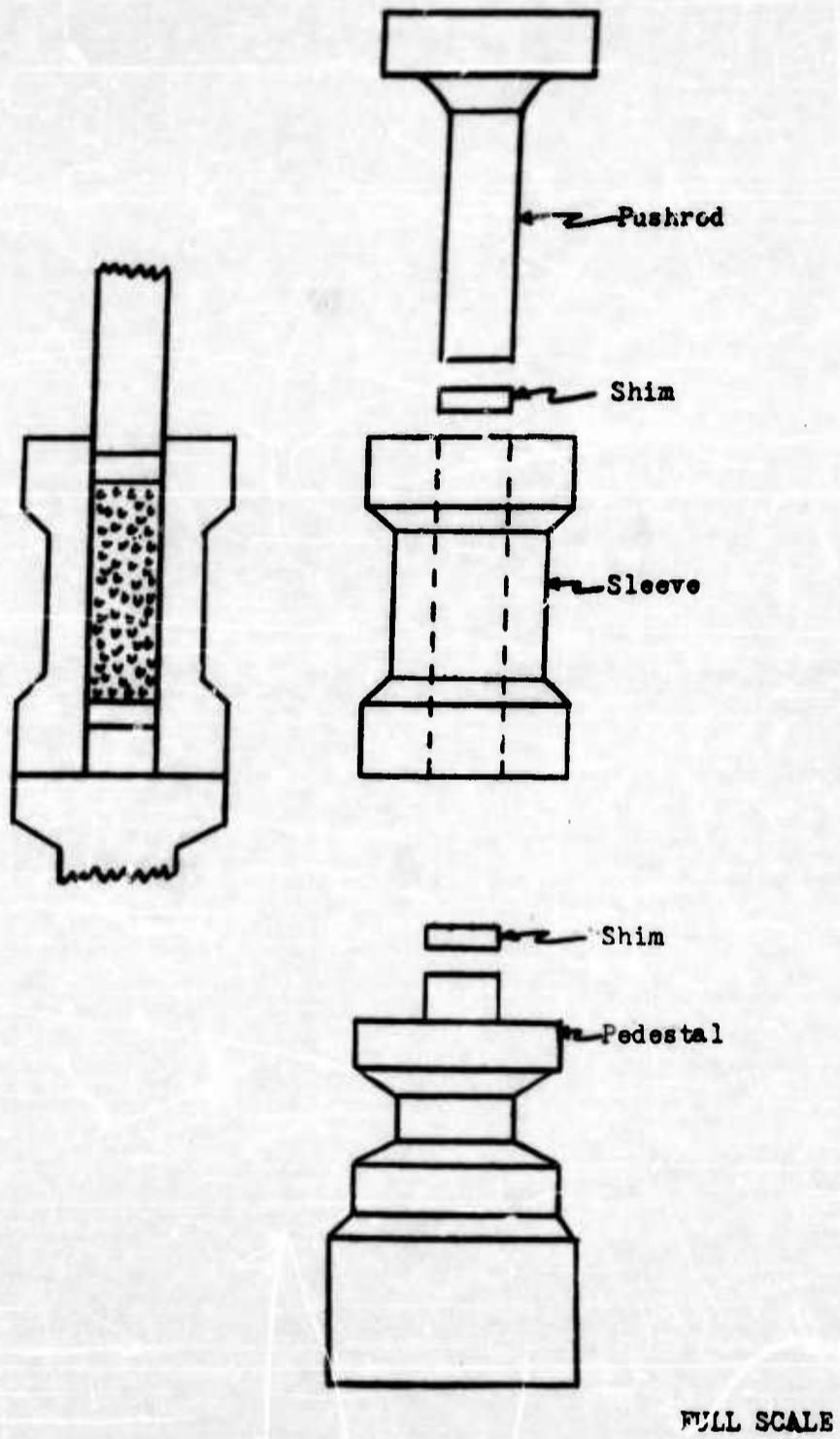


Fig. 1 Typical Die Assembly Used for Hot Pressing Cylindrical Samples by Resistance Heating

As shown in Fig. 2, the furnace is a water-cooled brass shell with a quarter-turn lock base, and a sliding plunger top, both of which contact the pressing surfaces of the graphite die assembly and effect the transmission of electric current through the system.

The complete hot-press unit is shown in Fig. 3. Pressure is applied to the die assembly by means of a lever system actuated by a hydraulic ram. The ram is powered by water under normal line pressure. Power to heat the graphite die is supplied through the top and bottom electrodes from a 50 KW transformer, of low secondary voltage. The amount of compaction of the hot-pressed material is indicated by the depression scale positioned near the ram assembly.

In a typical hot-pressing operation, a weighed amount of powdered material was placed in the graphite mold and compacted on an arbor press. The die assembly was placed in the furnace, the jacket was locked in place, and an initial pressure was applied to the die to effect satisfactory electrical contact. Cooling water was turned on for the furnace jacket, flushing gas was passed through the chamber, an optical pyrometer was positioned to sight on the die through the window and the power was turned on. Approximately 2000 amperes at 10 volts were used to attain the necessary temperature. When the appropriate temperature (as measured by the optical pyrometer sighting on the graphite sleeve surrounding the specimen) was reached, ram pressure was increased to provide a pressure of about 2000 psi on the specimen in the die. Pressure and temperature were held until the desired amount of depression of the specimen was achieved. Then, power and pressure were turned off and the

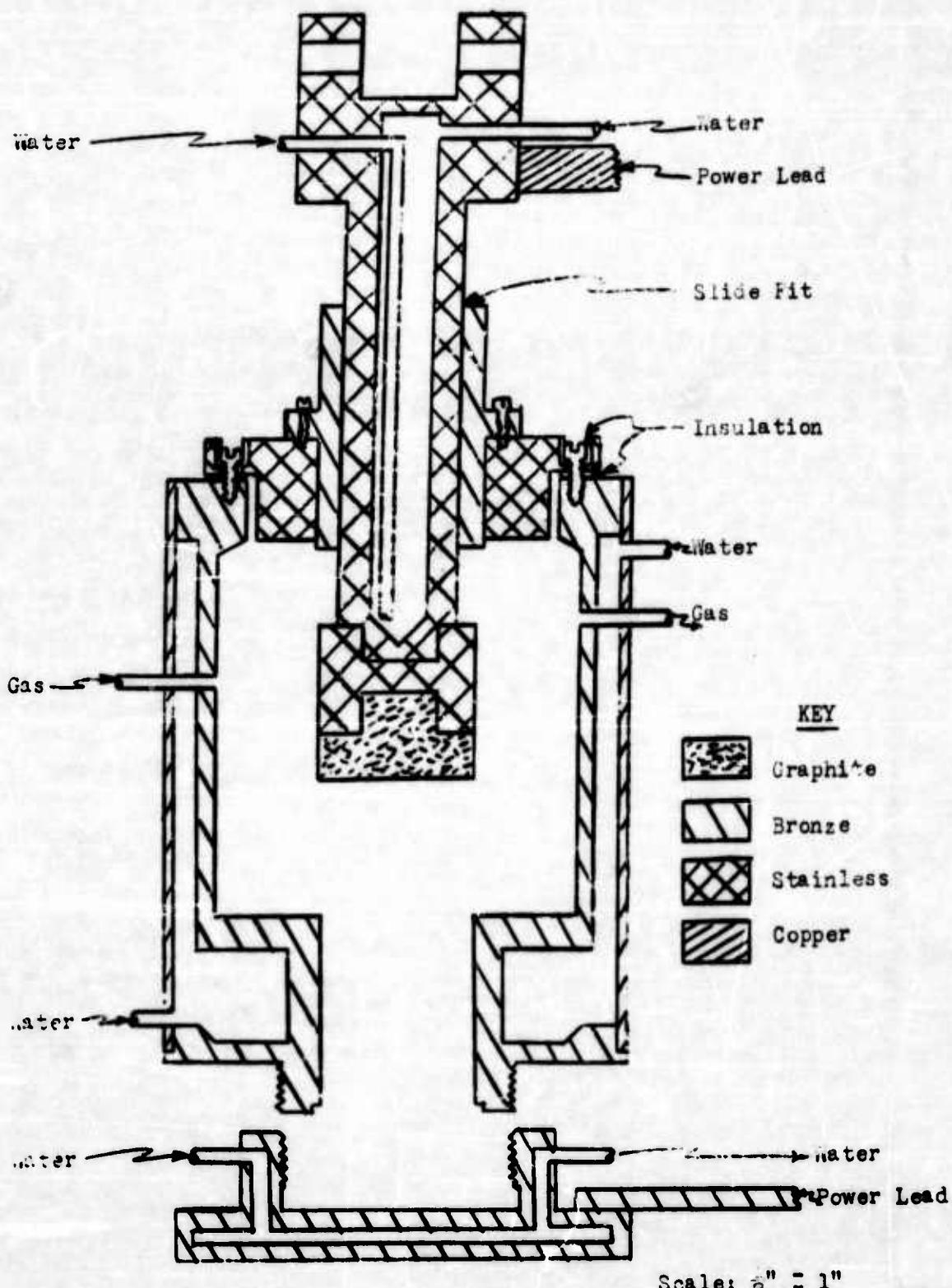


Fig. 2 JACKETED HOT PRESS FURNACE

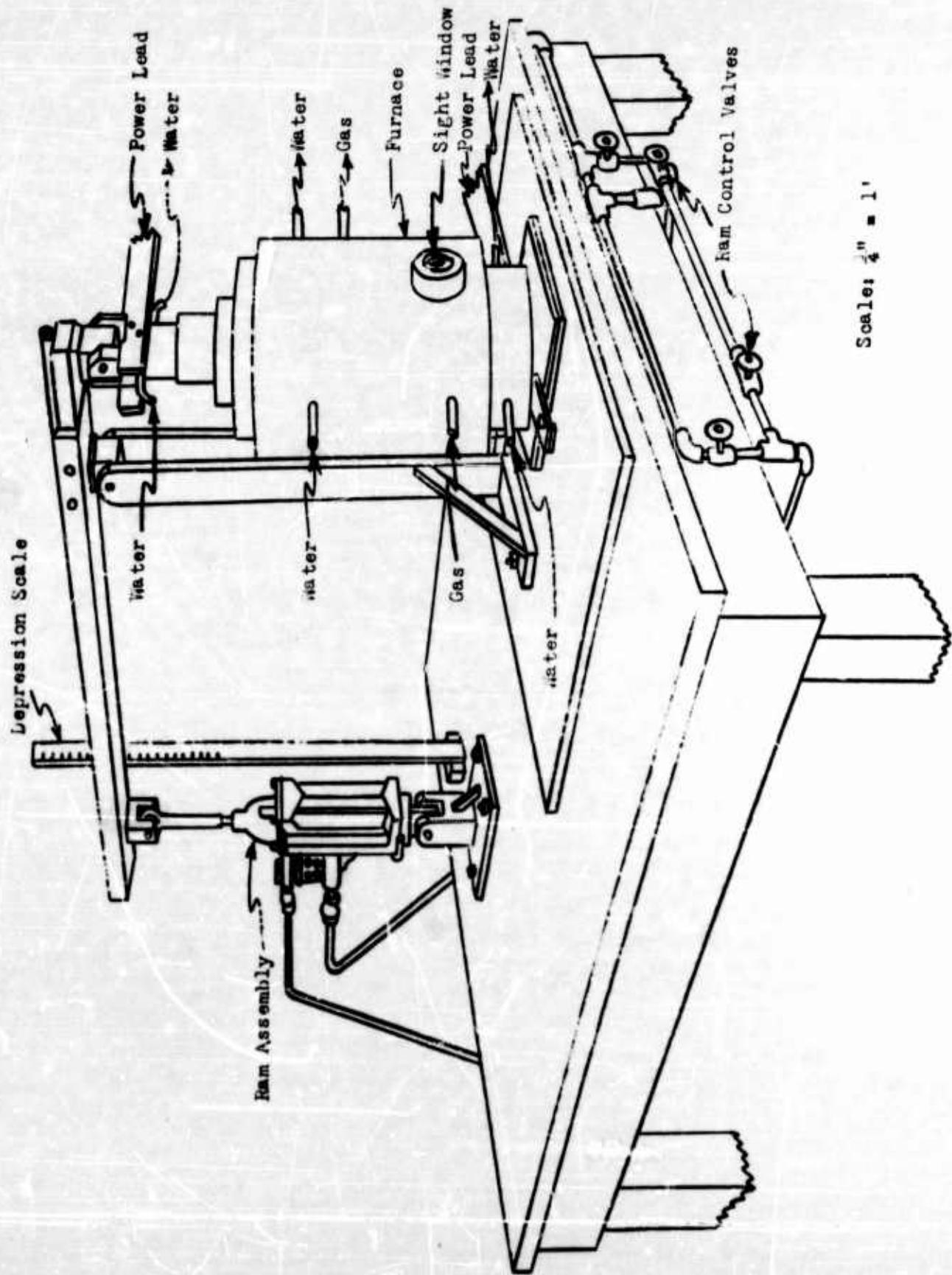


Fig. 3 Hot Press Furnace with Ram Assembly

die was allowed to cool in place. With this method, a maximum temperature of about 2500°C could be reached in about one and one-half minutes, and the complete cycle, including cooling, required only about ten minutes. Under the best conditions and with an adequate supply of graphite die parts, it was possible to hot press as many as six specimens in one hour.

The resistance hot press proved to be very successful, and many specimens were fabricated of intermetallic compositions including aluminides, borides, nitrides, carbides and silicides. Two-component and three-component systems of metals, intermetallics and oxides were also prepared by this technique.

A program to study container materials for melting titanium³ utilized this hot-pressing method. During a continuation of this work under different sponsorship⁴, it was necessary to replace the original furnace jacket assembly with a larger one which followed the original design; this revision permitted the use of larger graphite dies in order to hot press crucibles having a diameter of three-fourths inch.

The hot press was used to great advantage for Bachelor's theses^{5,6} studying the applicability of hot-pressed oxides for cutting tools. Highly densified oxides, with minor additions, were found to retain their small particle size when formed by resistance hot pressing.

B. Large Resistance Hot Press

In order to hot press larger specimens than were possible with the jacketed resistance hot press, and to achieve greater pressures, a seven and

one-half ton capacity hydraulic press unit was adapted for hot pressing by the addition of an electrode assembly. The complete assembly is pictured in Fig. 4; this shows the arrangement for a typical hot pressing operation, with insulation around the die and vertical push-rod in place to transmit pressure to the die from the movable press platen.

A schematic diagram of the resistance hot press is shown in Fig. 5. A graphite mold (c) is clamped horizontally between graphite electrodes (b) and is heated by virtue of its electrical resistance. Water-cooled copper heads (a) serve a two-fold purpose: 1) they prevent overheating of the electrical contacts during operation, and 2) they provide a rapid means for removing heat from the mold during the cooling cycle.

When all graphite parts are used between the electrodes (b) a severe temperature gradient occurs across the length of the mold. This is caused by the high thermal conductivity and mass of the graphite electrodes and electrical contacts, and the channeling of current around the mold cavity.

This problem was overcome by placing carbon "resistor" plates (e) between the mold and the large electrodes (b). The small graphite plates (f) serve only as protection for the electrodes (b). The electrical and thermal resistivity of a b.c.f. is considerably higher than that of graphite (see Appendix). Therefore the carbon plates serve in a two-fold manner to decrease the temperature gradient across the mold: 1) by virtue of their low thermal conductivity they provide a thermal barrier between the mold and the conducting end-members and 2) because of their higher electrical resistivity they dissipate more electrical energy as heat than does the graphite mold.

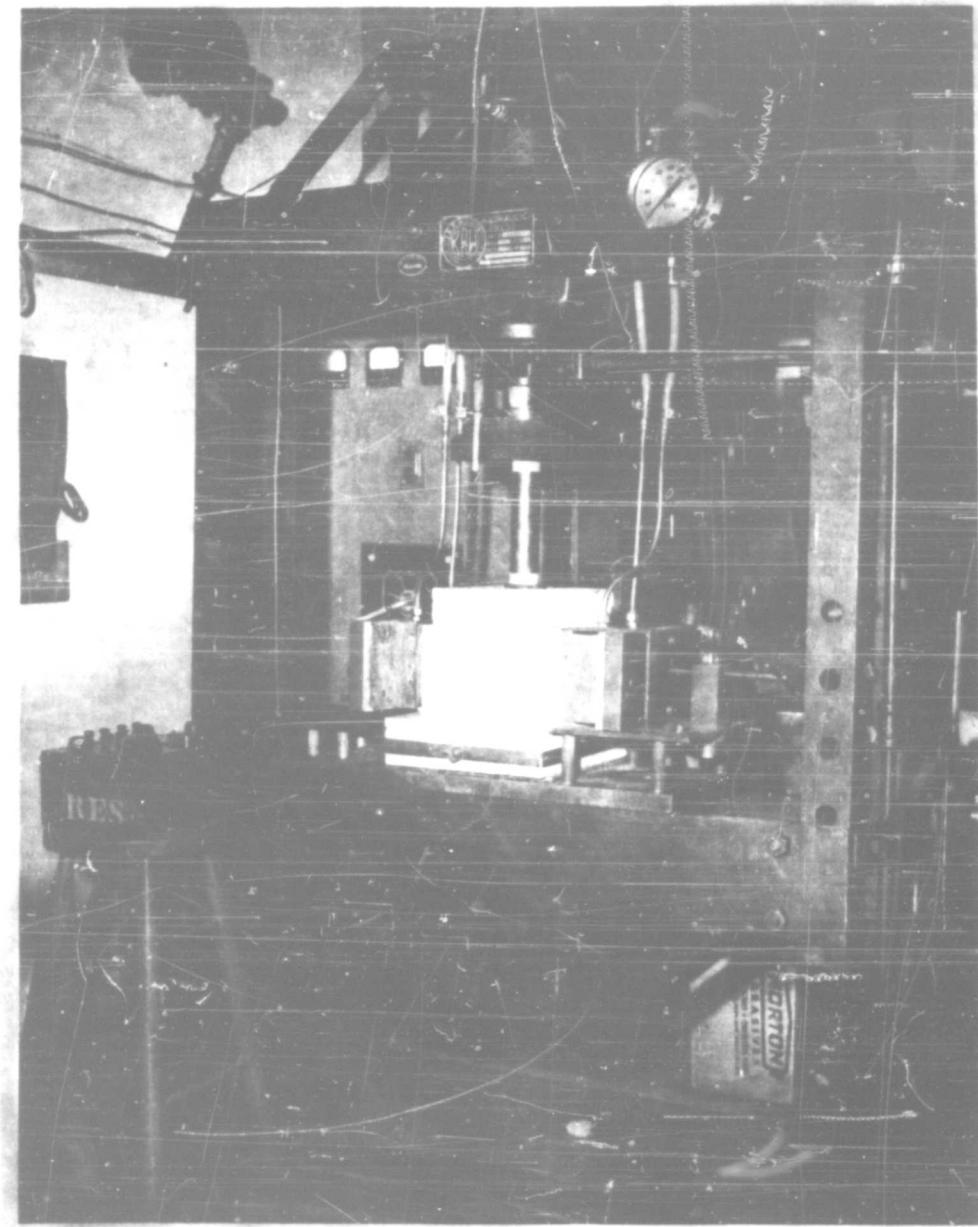


Fig. 4 Large Resistance Hot Press

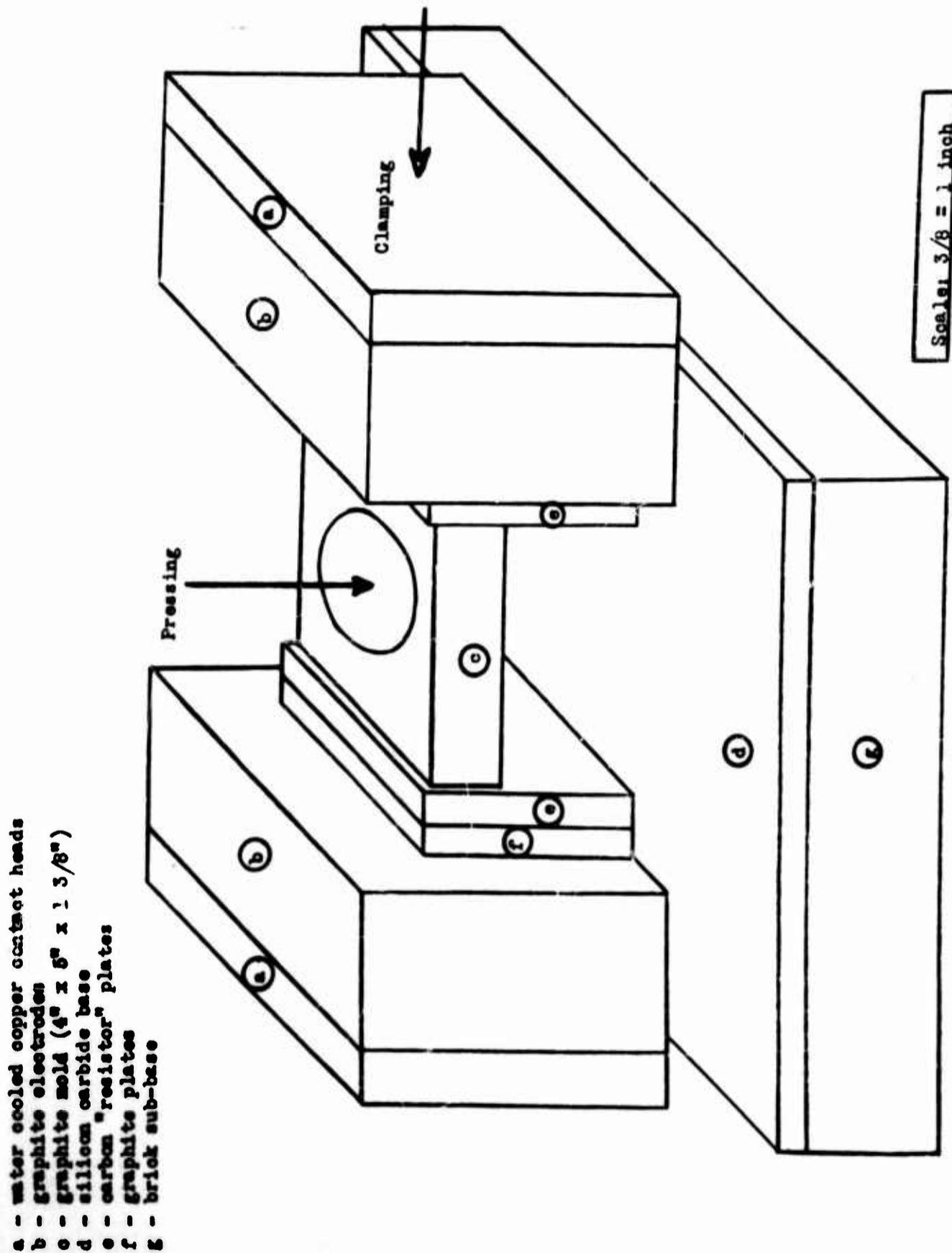


Fig. 5 Schematic Diagram of Resistant Hot Press
(copper electrodes and leads not shown)

The choice of a carbon "resistor" plate for this application depends upon the electrical and thermal properties of both the carbon and the graphite used. Since these properties vary considerably in these materials, it is simpler to select the proper thickness of carbon by trial and error methods.

Pressure is transmitted to the mold cavity through a cylindrical carbon rod. The lower push rod of the graphite mold is in turn supported by a cylindrical carbon base which extends through the lower insulation to the silicon carbide slab base. The present hydraulic system is manually operated; however, it is entirely feasible to incorporate a hydraulic pumping system if it were needed.

For many operations, temperature is measured by means of a Pt. vs. Pt. - 10% Rh. thermocouple inserted up through a hole in the carbon base-block, into a thermocouple well in the lower plunger of the die. A dense, high-alumina protection tube is used to prevent carbon contamination of the thermocouple. An optical pyrometer is used for higher temperature requirements; the pyrometer is positioned to sight through a graphite tube into a suitable cavity located in the upper side of the mold.

Power is supplied to the hot-press assembly by the 50 KW transformer operating at 230 V, 250 A maximum (Fig. 6). The ignitron contractor controls the primary circuit of the power transformer by means of the two ignitron tubes, connected in inverse parallel, which conduct the primary current. Firing of the ignitrons at the desired point on the supply-voltage cycle is accomplished by the heat control panel containing two thyratron tubes in inverse parallel. The effective current per cycle passed by the ignitrons is controlled by the phase-shift network of the heat control device. This control is effected by means of the

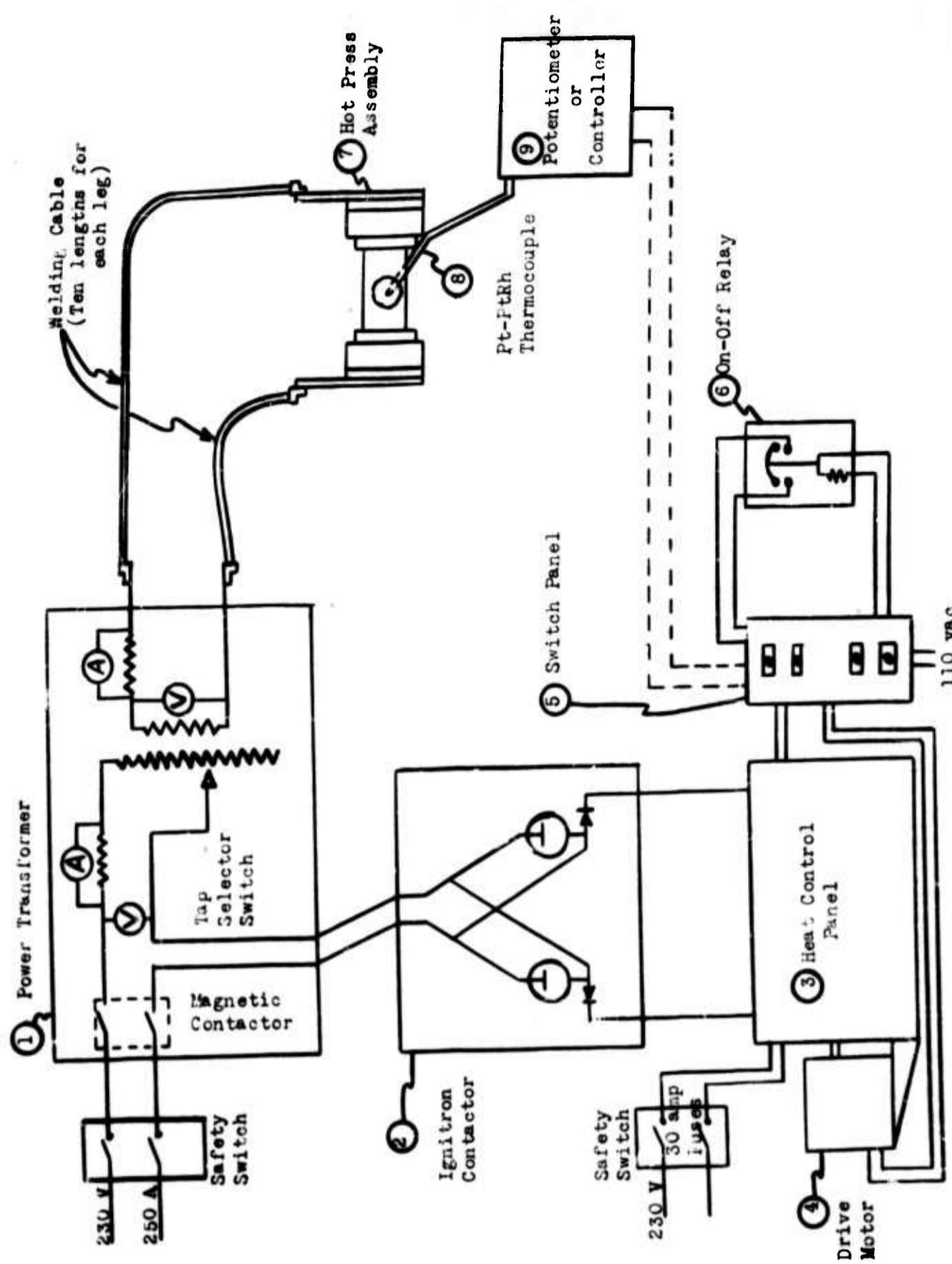


Fig. 6 Block Diagram of Power Control Circuit

potentiometer adjusted by the reversible control-drive motor. The motor is actuated by a manually-operated three-position toggle switch. The instrumentation of this circuitry is such as to permit automatic control of heating by connecting the thermocouple leads, or radiation pyrometer output, to an electronic controller which will actuate the control-drive motor.

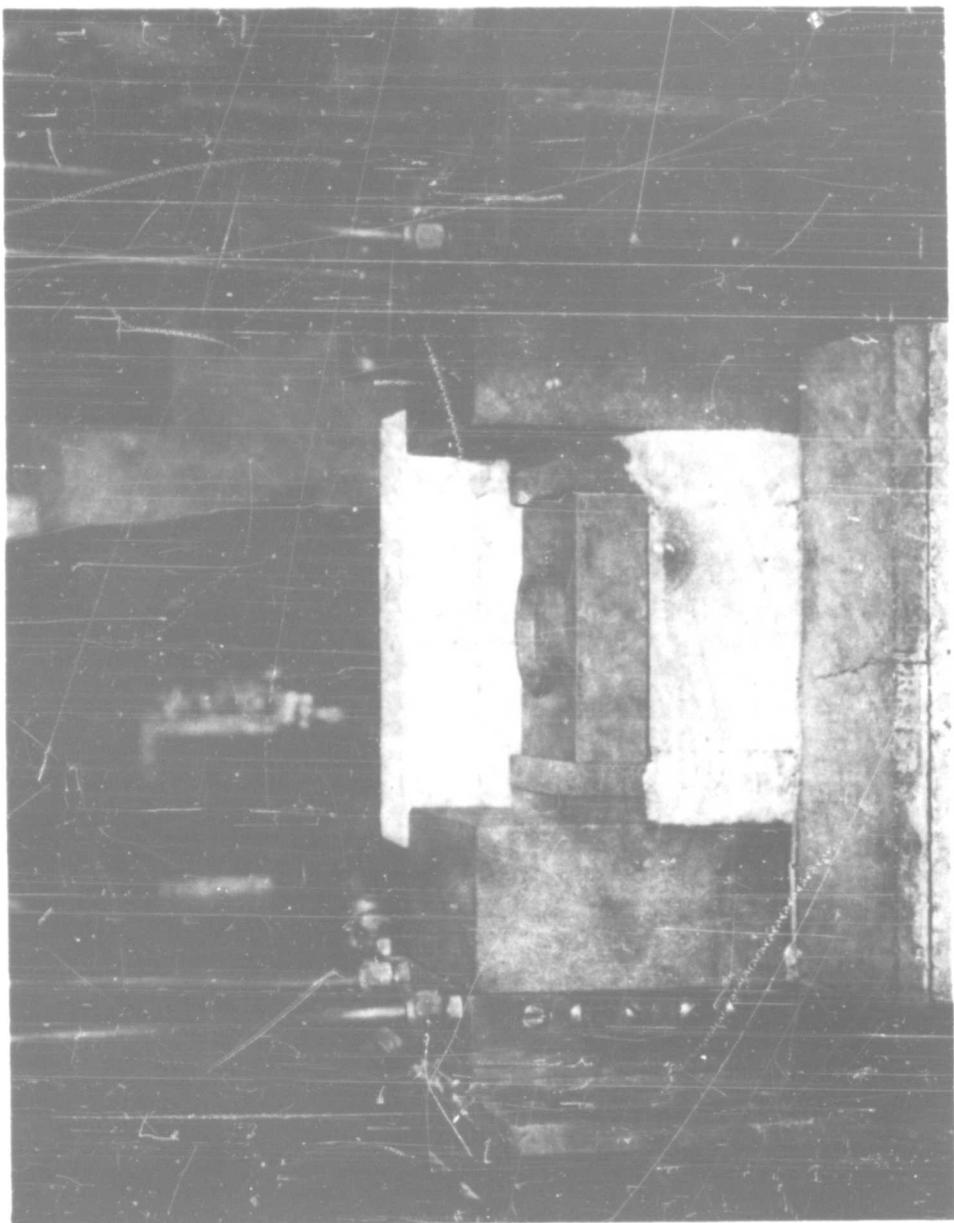
In order to prevent serious oxidation of the graphite parts, it is necessary to provide insulation which will act as a barrier to the infiltration of air. For temperatures up to about 1350°C , a tightly-packed layer of Fiberfrax surrounded by Alfrax brick is effective. Where high temperatures are to be encountered, MgO powder is used in direct contact with the die, then Fiberfrax and Alfraxbrick.

Fig. 7 shows a typical die clamped in place between the electrodes, before installation of side and top insulation. The same die is shown, in Fig. 8, in operation at about 1100°C , with the side insulation removed to illustrate the uniform temperature distribution achieved throughout the length of the die. With this set-up it is possible to form specimens of other than just cylindrical cross-section; four inch and five inch long bars are pressed regularly, as well as many special shapes.

Die washes of finely-powdered boron nitride or high-grade alumina are dispersed in an alcohol-water mixture and then painted on the graphite parts which constitute the die cavity. Such coatings are very effective in preventing reaction between the compact and the die parts.

An assembly much like that shown in Fig. 7 is used for a typical pressing of a metal-intermetallic mixture. The weighed charge is distributed uniformly in the coated die cavity, then the top plunger of the die is positioned carefully. The

Fig. 7 Resistance Hot Press Showing Interior with Mold Exposed



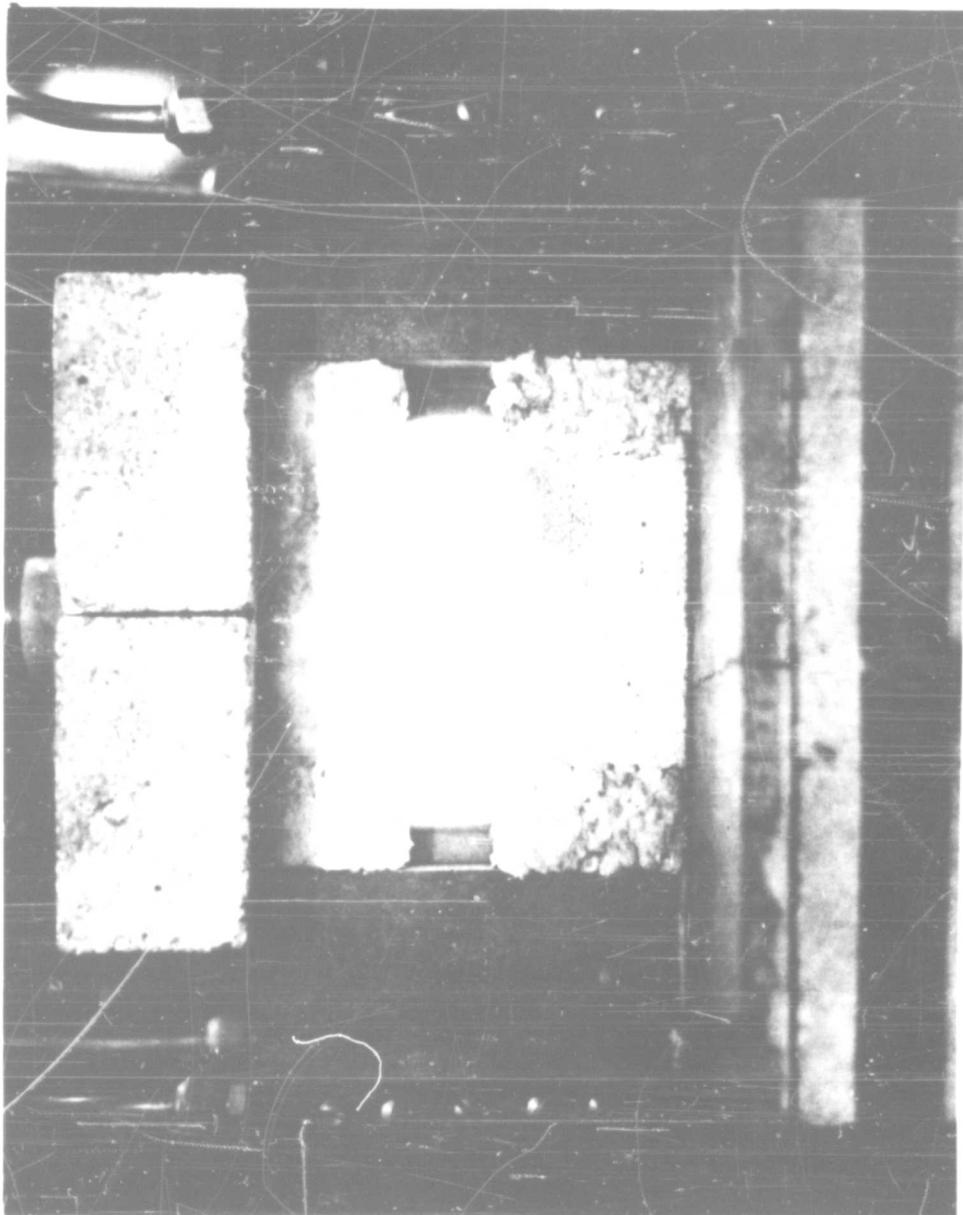


Fig. 8 Resistance Hot Press in Operation with Side Insulation Removed
(Mold Temperature 1100°C)

assembled die is put in an arbor press to pre-press the compact, after which the die is placed in the hot-press furnace. Uniform contact of the furnace plates against the end faces of the die is accomplished by adjustment of the clamping mechanism; it is important that this adjustment be made carefully to eliminate any tendency towards non-uniform heating occasioned by misalignment of the contacting surfaces. The carbon pushrod and the insulated steel extension rod are positioned to apply pressure to the top plunger of the die. Then, the Fiberfrax insulation is packed securely around the die, and the Alfrax brick are put in place.

Water is turned on for the water-cooled heads, and the power supply is put into operation. The initial reading of the slump gage (measuring the movement of the pressing platen) is taken. The desired temperature (1300°C , for example) is reached in about fifteen minutes under secondary output of 3000 amps at 5 to 6 volts. The proper pressure is applied during the final stages of heating until the desired compaction of the specimen, as indicated by the slump gage, is achieved. Then the power is shut off, and the furnace is allowed to cool to room temperature; this usually requires only 30 to 40 minutes. Under these conditions, and allowing ten minutes to make necessary adjustments in the initial positioning and clamping of the die, one pressing cycle per hour can be accomplished without difficulty.

One-, two- and three-component systems of metals, intermetallics and oxides have been successfully hot pressed in this resistance hot-press furnace; temperatures up to about 1850°C have been reached easily. Of particular interest is an investigation currently in progress in which rectangular bars of high-

purity alumina are being hot pressed with very limited grain growth occurring. This is attributed to the relatively short cycling time possible. In order to keep the grains down to near one micron particle size, ten minutes', or less, time is used for the high temperature portion of the heating cycle.

III. SUMMARY

(1) Resistance hot pressing offers a rapid method of forming specimens; this feature limits the possibility of grain growth, and permits fast production of samples.

(2) This technique makes possible the formation of a dense product composed of materials having widely differing melting points; for example, a cermet containing alumina (M. P. 2050°C) and nickel (M. P. 1452°C).

(3) Resistance hot pressing is advantageous for fabricating mixtures containing macro-sizes of metallic phases which would be subject to hysteresis loss heating effects in induction hot pressing.

(4) This method will produce dense specimens of materials which cannot be sintered normally because of decomposition tendencies.

(5) When normal precautions are observed in loading and positioning the die, uniform high density products may be produced.

(6) The use of graphite dies permits the design and fabrication of special shapes, in addition to the usual cylindrical shapes.

(7) Temperature measurement and control is readily effected with the resistance hot press.

↓
⑧ The costs of constructing and operating a resistance hot press are very reasonable because of the simplicity of design and the nominal power supply requirements.
↑

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G. Potter

J. B. Schultz

M. A. Tuttle.

APPENDIX

The Selection and Use of Carbon and Graphite in the Design of the Resistance Hot Press

One of the most serious problems encountered in a resistance hot-press design is the provision for a uniform temperature zone in and around the specimen. To a large extent this problem can be overcome in small-sample fabrication by using cylindrical mold bodies with reduced cross sections and by allowing the current to flow parallel to the cylinder axis.

The problem becomes more acute, however, when attempting to hot press large, intricately-shaped specimens such as cylinders, rings and bars. It is sometimes physically impossible, and always expensive, to contour large graphite molds in order to produce a uniform distribution of current flow and temperature around the specimen.

In many cases it is possible to overcome this difficulty in large hot-pressing operations by the utilization of both carbon and graphite materials in a furnace design. The properties of graphite as a resistance-heating material are well known. Carbon is not used normally in applications of this type because of its much higher electrical resistivity and its higher hardness, which makes it difficult to machine. Carbon also has a much lower thermal conductivity than does graphite.

These two properties, higher electrical resistivity and lower thermal conductivity as compared to graphite, make carbon an ideal material for use in a resistance heating application.

The application described on page 9 of this report is a very good example illustrating the use of carbon in a hot-pressing operation. The carbon "resistor"

plates which were placed between the mold body and the conducting graphite electrodes (Fig. 5) produced the following effects:

1) by virtue of their higher electrical resistivity, the carbon plates dissipate more electrical energy as heat than does the smallest cross-section of the graphite mold;

2) because of their lower thermal conductivity, the carbon plates act as thermal barriers to retard the flow of heat from the mold into the large conducting graphite electrodes.

In combination, the effects noted above provide for a uniform temperature distribution over the entire length of the mold (Fig. 8). In this application, a uniform temperature zone was obtained around the specimen without resorting to a large, intricately-shaped, expensive mold body.

The use of carbon in conjunction with graphite in a furnace design is not limited to the application mentioned above. Carbon can be used in any furnace where it is desirable to control or limit the flow of heat or current, or both. Another example demonstrating the versatility of carbon in furnace design was indicated by Tinklepaugh¹. Carbon blocks are used in the upper and lower ends of induction hot presses to transmit pressure and to act as thermal barriers to the flow of heat into the water-cooled heads and bases. The high electrical resistivity of carbon is also an advantage in this application in that it is less efficient as a susceptor than graphite, and therefore contributes to concentrating the heating in the mold section.

Some of the advantages derived from the use of carbon in conjunction with graphite in a resistance hot-press design include the following:

- 1) will provide temperature uniformity over larger mold areas than could otherwise be obtained with graphite alone;
- 2) furnace and element (mold) size can be kept to a minimum, thereby reducing the initial furnace and mold costs;
- 3) smaller furnace mass allows for more rapid cycling (this is a very important consideration when working with very fine materials which exhibit rapid grain growth on prolonged exposure to elevated temperatures);
- 4) simple, easily-machinable molds and furnace parts can be used;
- 5) carbon incorporated in the furnace design makes the resistance hot press far more versatile with respect to the power supply (it is possible to adjust the resistance of the furnace, independent of the mold size or shape, by proper selection of carbon parts).

Graphite, because of its electrical properties and stability at elevated temperatures, is an ideal mold material for resistance hot-pressing applications. Although not essentially a strong structural material, graphite is unique in that it increases in strength with temperature up to about 2400°C . There are few readily available materials stronger than graphite at temperatures in excess of 1600°C . However, carbon and graphite parts must be protected from atmospheric oxygen at temperatures above 600°C .

The increase in strength, with temperature, of graphite has been explained by the internal stresses in the polycrystalline material which result from anisotropy of thermal expansion². It has also been pointed out^{2,3} that, since the pores in artificial graphite provide stress concentrations, some relaxation of stress can occur at temperatures high enough for grain boundary flow. Therefore, a greater average stress is required to produce fracture at higher temperatures than at lower temperatures.

Because of the anisotropic nature of artificial graphite, preferred orientation during forming gives rise to considerable variation in physical properties. This is shown in Table I wherein the physical properties of a fine-grained molded graphite are compared with those of a similar graphite formed by extrusion. It is apparent from these data that extrusion produces a greater degree of anisotropy than does molding.

TABLE I⁴
Effect of Molding vs. Extrusion on the
Physical Properties of a Fine-grained Stock

	Extruded stock 1.64	Molded stock 1.75
Density		
Specific Resistance (Milliohms-cm)		
Parallel to grain	0.86	0.96
Perpendicular to grain	1.62	1.32
Coefficient of Thermal Expansion ($\times 10^{-7} / {}^{\circ}\text{C}$)		
Parallel to grain	11	19
Perpendicular to grain	41	32
Elastic Modulus (10^6 lb/in^2)		
Parallel to grain	1.84	1.39
Perpendicular to grain	0.78	0.96
Flexural Strength (lb/in^2)		
Parallel to grain	4520	4680
Perpendicular to grain	3010	3940

The selection of a graphite stock for resistance hot-pressing molds and furnace parts requires some knowledge and understanding of the anisotropic nature of the physical properties of the material. Many of the less expensive, extruded, electrode-grade graphites are suitable as mold stock for some pressing operations, but in general these grades are characterised by laminations and non-uniform structures. It is difficult to select, from those stocks, good mold blanks which will withstand stress levels that are average for that particular grade.

The molded grades of graphite are much more uniform in this respect, and in general they make better molds and parts for resistance hot pressing.

New forms of carbon and graphite, produced by molding and multiple impregnation techniques, are now available from several manufacturers. These materials are more isotropic with respect to their physical properties and are characterized by high density and strength values. Compressive and tensile strength values have been reported in the ranges 8000 to 10,000 psi, and 2000 to 3000 psi, respectively. These new graphite products look very promising as mold and structural materials for resistance hot-pressing furnace design.

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